1965

Horizontal distillation

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HORIZONTAL DISTILLATION

by

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ABSTRACT

A horizontal distillation device was developed to provide a good separation at low pressure drop. The device developed was a hollow tube three inches square and eight feet long. The horizontally placed tube was heated along the bottom side and cooled along the top. The heat was supplied by electric heaters and the cooling medium was water. A conventional reboiler was at one end of the tube and a condenser at the opposite end.

The distillation device was tested using the acetone-methanol system. It was successfully operated under conditions of total and partial reflux. At total reflux the maximum number of theoretical stages observed was 3.5. A total pressure drop of less than 5 mm water was observed at all operating conditions tested. The separation achieved was better with the tube in a horizontal position rather than inclined 3° to the reboiler end. Maximum separation was observed when the heat input-heat output ratio of the tube approached one.

A mathematical model was developed to express the liquid composition in the tube as a function of the position in the tube. The model fits the observed liquid composition profile within experimental error. The number of stages calculated by the model was in good agreement with the number of stages calculated by the McCabe-Thiele method.

INTRODUCTION

Much theoretical and practical work has been done in the area of continuous distillation. Most of this work has been done on conventional packed columns and bubble cap and sieve tray columns. Since the time of the invention of the bubble cap column, very little work has been done to find new methods and techniques for the separation of materials based on boiling point differences.

A horizontal fractionator is proposed to effect a good separation at low pressure drop with reasonable capacity. Because vapor flow is restricted and a liquid seal is required, bubble cap and sieve tray columns operate at high capacities with relatively high pressure drop. The use of packed columns to reduce pressure drop has not been successful in large sizes because of efficiency losses from channeling.

The method of achieving the separation is different in a horizontal unit than in a conventional tray or packed column. In a bubble cap or sieve tray column the separation is achieved by intimate gas-liquid contact on the tray. In a packed column this vapor-liquid contacting is achieved by dispersing the liquid over the packing to provide a large area for the vapor to contact the liquid. In the horizontal unit studied the separation is not dependent primarily on intimate vapor-liquid contact. It is dependent on

continuously boiling and condensing the liquid and vapor streams.

Distillation has been accomplished in a horizontal hollow tube (2, 7). The separation is accomplished by heating the liquid phase, on the bottom of the tube, and cooling the vapor phase on the top of the tube. The net vapor velocity from the reboiler through the tube to the condenser effects the vapor-liquid contacting. Since the tube is hollow, there are no expensive tray structures or packing on the inside of the tube. Therefore, the pressure drop is very low.

If a horizontal distillation unit of this type can be made commerically feasible, it would have several advantages over conventional distillation columns. Because of its very low pressure drop, distillation of heat sensistive materials could be carried out economically at low pressures. Since there are no expensive tray devices or packing the first cost of such a unit should be very low. This could also reduce the maintenance cost. The cost of construction should be reduced because the unit should take less structrual support than a conventional vertical column.

The horizontal device used in this study was a hollow, square tube. The area of interest was to study the heating and cooling variables to determine the

operating conditions. The effect of the slope of the tube was also investigated. A mathematical model was proposed to express the liquid composition profile in the tube.

LITERATURE SURVEY

Horizontal distillation is not new. It is closely associated with the history of distillation. Forbes (3), in his excellent book on the history of distillation, indicates that horizontal distillation units were used before the conventional vertical units.

Distillation was always done in a simple pot until about 1800. The only significant contribution before that was the use of running water in the condenser about 1275. Modern distillation theory had to wait until the theories of heat and vaporization were developed around the middle of the 18th century.

The first multistage column was built in 1801 in France by Edouard Adam. The Adam still was three Woulfe bottles connected in such a way that the vapors from each were piped to the others. This was the first instance of letting the rising vapors from the still meet the returning liquid to effect the distillation. Each of the bottles was heated individually, but only air was used as the cooling medium in the condensers. The Adam still and its modifications were popular for about ten years.

Isaac Berard was the inventor of a still of greater simplicity in 1804. The vapors from this still passed through cylindrical tubes which were divided into compartments

by perforated plates. The condensing liquid was conducted back to the still. The tubes were run horizontally. Berard used the principle of partial condensing rather than the individual reboilers used by Adam. The Berard still did not have the heat economy that was present in the Adam still, but was much simpler to operate. Hence, the Berard still was very popular until about 1840.

Because of the popularity of the Berard still, it is only natural that many imitators were hard at work. Two of the most important were Augustin Menard and Pierre Alegre. The Menard still, invented in 1804, was a horizontal device consisting of several compartments. Each compartment was a Woulfe bottle. The condensate from the bottle was not led to the next one but back to the still. Although this was a step backwards, as it gave up the counter-current flow ideas advanced by Adam and Berard, it did give Alegre his ideas.

The Alegre still, invented in 1806, tried to combine the ideas of Adam, Berard and Menard. In this horizontal apparatus the vapors from the still pass through a hermetically sealed Woulfe bottle into a series of Menard compartments, which act as partial condensers. The vapors then go to a preheater and then to a final worm cooler. This apparatus worked fairly well, but was too complicated to operate very efficiently.

Thus, in a matter of four years the idea of counter-current operation was developed and widely accepted. These men used the liquid to absorb the heat of the vapors and condense them, thus effecting better heat economy. The main drawback of these devices was that they were batch operations, and they would only distill the low viscosity liquids of the French wine producers.

An appartus to distill continuously was invented in 1808 by Jean Cellier. This device was used to distill sugar beet mash. The invention of Cellier was probably the most important contribution by one man to the science of distillation. The apparatus was very simple, it consisted of a column, derived from the Alegre and Menard stills, with bubble caps mounted vertically over the pot. This still was very close to the design of most distillation devices in use today. Cellier also invented sieve trays to be used with low viscosity liquids. Pierre Savalle was a pupil of Celliers and in 1816 and 1817 patented a better column with preheaters and total condensers. In 1824 another pupil of Celliers, A. P. Dubrunfaut, wrote the first text on distillation calculations and classifications.

Although Celliers column was very good, it was slow to be accepted. By the 1820's, however, it was in wide use and the arguments were centered around how

many bubble caps to put on a tray and how far apart to put the trays. The arguments do not sound much different from todays.

Horizontal distillation devices enjoyed wide popularity from 1801 to about 1830. Then, with the invention of Cellier's column, almost all research was stopped on different types of distillation devices until the 1950's.

In 1953 Rollet (7) introduced a horizontal fractionation device. The column is inclined slightly from the horizontal. It contains no packing or other restrictions to vapor flow. The bottom of the column is heated and the top of the column cooled. A reboiler is at one end, a condenser at the other. The column is inclined so that the liquid returns from the column to the reboiler.

During the operation of the column, the bottom liquid is boiling and its vapors mix with the vapors from the reboiler. A part of this vapor is condensed and falls back to the boiling liquid. These exchanges are multiplied the entire length of the column. Thus rectification of the liquid is accomplished.

The Rollet column was 0.76 cm, inside diameter, and 100 cm long. A bed of fritted glass, 2 to 3 mm wide, was laid along the bottom of the column to serve as the heating area. The heating along the column was accomplished by a bare nichrome wire connected to a variable voltage transformer. The cooling device along the top of the column

consisted of three copper tubes mounted parallel to each other. A sheet of copper, plated on the pyrex tube, made it possible to cool the upper portion of the column. Water was the cooling medium. A large glass tube enclosed the entire column. The reboiler was a 300 cubic centimeter flask, insulated and heated electrically.

The benzene-dichlorethane system was used for Rollet's studies. A slope of 4.5% was used for the majority of the experiments. Some studies were made at slopes of 1% and 9%. The increase in slope did not change the efficiency of the column. However, the change in slope did affect the time to reach equilibrum. The time varied from 1 hour at 9% to 3 hours at 1% slope. At the 4½% slope the liquid hold up was between 3 and 4 centimeters. The vapor velocity was about 1.95 centimeters per second. The number of theoretical plates was calculated from the Fenske equation. At a slope of 4.5% and at a total reflux rate of 2.20 cubic centimeters per hour (flow returning to the column) the number of stages was 28. If 16 cubic centimeters per hour were withdrawn the number of stages reduced to 9. No pressure drop data was recorded.

A horizontal distillation unit was patented by Andrew Spence (8) in 1955. This unit was used for the distillation of tall oil. This device consists of a long, narrow horizontal evaporator. This evaporator is heated

along its entire bottom side. A number of packed columns are placed on top of the evaporator. A condenser is placed on the top of each column and connected to a common vacuum header. The vapor section of the evaporator is divided into compartments. Part of the liquid from the condenser is fed into the evaporator section just proceeding it. Thus, the operation is similar to a number of packed towers (each used as one stage) set side by side using the same reboiler (evaporator). In this manner it is claimed that a very small pressure drop is measured from the evaporator to the condenser.

In 1959 Markels and Drew (5) introduced a semi-packed horizontal fractionation device. In this fractionator, the vapor flows straight through a horizontal duct, counter-current to the liquid which is pumped over a series of porous bats which act as cross-flow contactors. These bats may be set at angles of 55°, 67°, and 79° from the horizontal. The bats were made of knitted stainless steel forming a matrix of about 98% open volume.

The vapor efficiency was measured by humidifying the air, and liquid phase efficiency was measured by oxygen desorption from water by air. The two film efficiencies were combined to obtain an overall efficiency.

At an angle of 67°, liquid flow rate of 10.6 gpm/ft and vapor velocity of about 6 ft/sec, the pressure drop across the bat was 0.7 inches of water. The overall Murphee efficiency was calculated to be about 0.85.

The Eckey Horizontal Evapo-Fractionator (4) was introduced in 1963 by the Vulcan Manufacturing Company. The fractionator consists of a horizontal or slightly inclined cylindrical vessel containing a series of impellers mounted on a common shaft. The shaft is directly below and parallel to the axis of the shell. The impellers, which have a diameter about half that of the vessel, are almost entirely in the bottom half, leaving a large area above for the vapors to pass through. A series of compartments formed by partitions or dams about one-sixth as tall as the diameter of the vessel are located along the bottom. The liquid entering the eye of the impeller is sprayed into fine droplets directly across the open space above. A series of grids positioned on the upper part of the tank confines the liquid on the wall to its own compartment. As the coalesced liquid flows back to its own compartment, it is partially obstructed by a trough surrounding the impeller and divides into two streams. Part of this liquid returns to the front of the trough and blends with the liquid entering from the upstream compartment.

The remainder goes to the down stream side and then on to the next compartment. A column for processing 10,000 pounds per hour of oil would have 25 impellers. The fractionator is claimed to operate at less than 10% of the pressure drop of conventional vertical columns under comparable conditions. Typical pressure drop is in the range of 0.2 mm mercury per theoretical plate depending upon throughput.

Eberlin (2) reported work done on a Rollet type device in 1963. The Eberlin column was one meter long and 15 millimeters in diameter. The column was constructed of glass. The tube was heated with an electric heating wire running parallel to the axis of the tube. A cooling spiral was provided inside the tube for condensation. Boiling retardation was prevented by a chain of ceramic rings. The benzene-ethylene chloride system was used to determine the separation capabilities of the equipment. Variations in the vapor velocity, temperature of the cooling medium and the heat input to the tube were made to determine the separation capacity. A maximum of 12 theoretical stages was observed. The pressure drop was approximately 10 mm water over the tube.

EXPERIMENTAL

Apparatus and System

The purpose of this investigation was to obtain experimental data to determine the operating conditions of a horizontal distillation unit. The unit was patterned after the Rollet (7) device. It was felt that this device would have more practical value than the other horizontal devices. This was because it had no packing, baffles or mechanical devices to effect vapor liquid contacting. It was also felt that this device could be constructed economically.

The device consisted of a long hollow tube with a reboiler at one end and a condenser at the other. The tube was heated along the bottom and cooled along the top. The separation was carried out in the following manner. The vapor from the reboiler was condensed along the cold top surface of the tube. The condensed vapor fell, in droplet form, to the liquid pool along the bottom of the tube. This liquid was vaporized by the heat input to the bottom of the tube. This process, called internal reflux, was repeated many times as the vapor and liquid streams passed counter-currently through the tube. The enriched vapor was collected through the condenser. Part of this stream was returned as external reflux.

Several arbitary decisions were required before construction could begin. Because no information was available on the nature of the internal reflux, there was no way to know how much free vapor space should be available in the tube. Also there was no information on how long the tube should be. Therefore, it was decided to make the tube three inches square and eight feet long. The square tube was used for ease of assembly. The length was selected to be long enough to minimize end effects.

The tube was constructed from mild steel. The sides were made from three inch steel channels. Along one side were four locations where the feed stream could be introduced. Along the other side were located ten liquid sample ports, eight vapor sample ports and two positions to measure the pressure drop. The samples were taken with a hypodermic syringe. The top and bottom of the tube were bolted to the side channels. Synthetic rubber gaskets were used throughout the device.

The bottom of the tube was a three-sixteenth inch thick steel plate. To this steel plate four electric heaters were strapped. Each heater was 24 inches long and $2\frac{1}{2}$ inches wide. The heaters were a special design to allow very high heat fluxes, twenty five watts per square inch. Therefore, each heater had a capacity of

3100 watts. Each heater was controlled by a separate variable voltage transformer. The heaters were insulated from the air by three inches of fiberglass.

The top of the column was a three-sixteenth inch steel plate with sides one and one-half inches high. This formed an open channel along the top for the cooling water. The bottom of the top plate was roughed to allow sites where droplets formed and fell back to the boiling liquid on the bottom plate.

The reboiler was a 22,000 ml pyrex flask with a 2050 watt heating mantle. Each of the three heaters was controlled by a variable voltage transformer. The maximum boil up using the acetone-methanol system was estimated to be about three gallons per hour. The connection from the reboiler to the tube was a pyrex pipe. A calibrated siphon bottle was used to measure the flow returning to the reboiler. Provision was also made to introduce feed into the reboiler and also to remove the bottoms product through a cooler.

The condenser was a Grahm Model 6379 tube and shell heat exchanger with approximately $1\frac{1}{4}$ square feet of area. The vapors from the tube were condensed on the shell side and fell by gravity to a glass jar which was the accumulator. The cooling medium used on the tube

side was water. The condensate was pumped through a rotameter to a stream splitter which sent back the desired fraction to the tube with the remaining product drained to a storage drum. This reflux stream could be preheated before entering the tube by use of an electric heating tape about the pipe.

It was felt that the tilt from the horizontal position could be an important factor, as this angle determines the rate of the liquid return from the tube to the reboiler. To allow for changing this slope, it was decided to mount the tube, reboiler, condenser and accumulator on a platform that could be tilted. The above components were mounted on a two by twelve foot board, pivoted at the center with two hydraulic jacks at each end to adjust the slope.

Figure 1 is a schematic drawing of the completed device. The feed, bottoms and product tanks were thirty gallon stainless steel drums. All tubing was 316 stainless steel.

The system used in this study was acetone-methanol. The equilibrum diagram from Chu (1, p. 28) for this system is shown in Figure 2. This system was chosen for ease of analysis, by refractive index, and because it takes a number of stages at total reflux to obtain a large separation. It was also an economical system to use.

Figure 1. Schematic of the apparatus

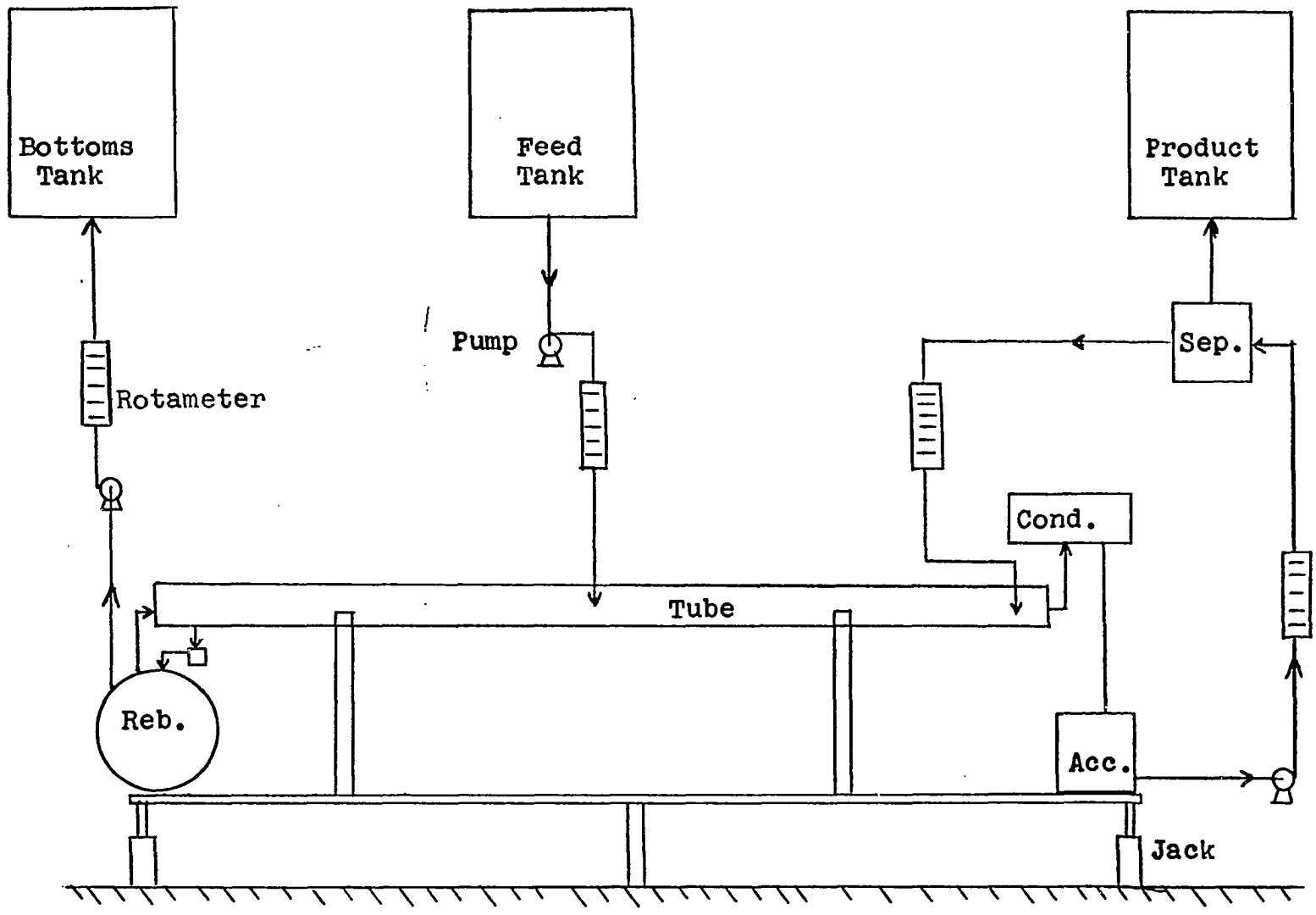
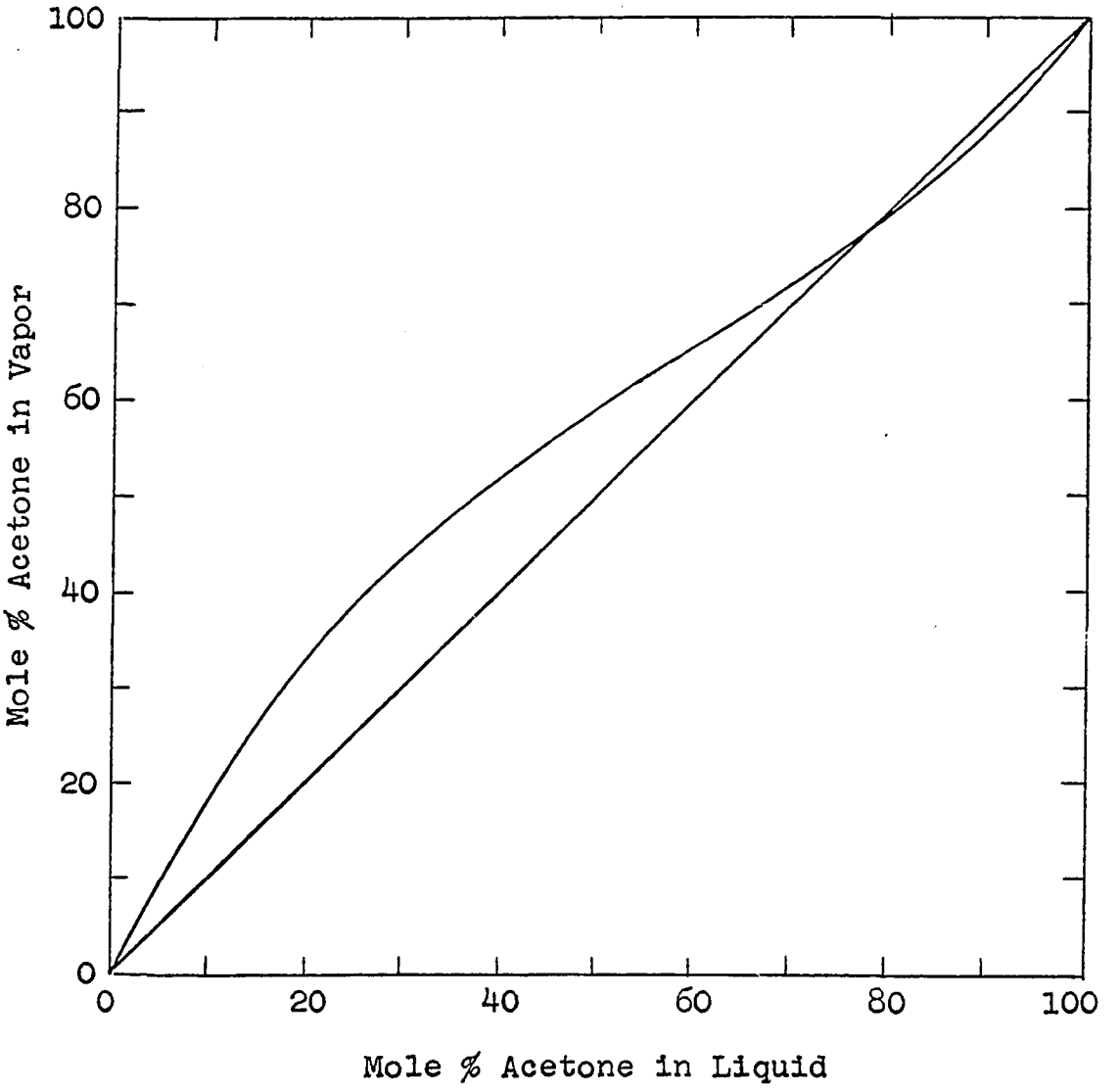

Bottoms Tank
Feed Tank
Product Tank
Pump
Sep.
Rotameter
Cond.
Tube
Reb.
Acc.
Jack

Figure 2. Acetone-methanol vapor-liquid equilibrum curve


100
80
60
40
20
0
0
20
40
60
80
100
Mole % Acetone in Vapor
Mole % Acetone in Liquid

The disadvantages of the system were the azeotrope at 80 mole per cent acetone and the non-constant relative volatility.

Procedure

A typical distillation run began with the preparation of a 20-25 mole percent acetone solution. This feed was then pumped into the reboiler until it was approximately half full. The reboiler was then started up at the desired heat input level. When the reboiler was boiling well, the heaters along the bottom of the tube were started and the cooling water was introduced to the top of the tube. It was important to start the cooling water and the heating at the same time, otherwise the heat would deform the column.

The condensate began to flow into the accumulator in about thirty minutes. The feed pump was started and the flow to the desired feed location was adjusted. The bottoms withdrawal and product removal rates were adjusted to maintain the material balance.

The operation was kept going for another one hour and thirty minutes at which time samples were withdrawn and analyzed. After another 15 minutes samples were again taken and compared to the first set. If no change was noted, it was assumed that the apparatus was

at steady state. If the unit was at steady state all data were recorded and the apparatus allowed to cool. Some runs were made back-to-back allowing two hours between sampling times.

In order to evaluate the capabilities of the fractionator the tilt, cooling water rate to the tube, and the heat input rate to the tube were varied. Two levels of each were selected. Runs were also made at total and partial external reflux. Evaluation of conventional distillation columns is often made at total extermal reflux, therefore, many of these runs were duplicated to check the reproducibility of the data.

RESULTS

Treatment of Data

The two important variables that determine the separation in a horizontal distillation device were the heat input to the tube and the heat removed from the tube by the cooling medium. Attempts were made to measure these requirements as accurately as possible.

A series of runs were made using pure acetone to determine the heat flux along the bottom of the tube as a function of the heater voltage. For these runs the tube was modified by disconnecting the reboiler and allowing the liquid along the bottom of the tube to flow into an external vessel. The tilt was adjusted so that the reboiler end was higher than the condenser end. No cooling water was used and the top of the tube was insulated to minimize condensation. The acetone was fed to the condenser end of the tube. The vapor resulting from boiling along the tube was condensed in the column condenser and collected. Material and energy balances were made to determine the heat flux at the various heater voltages. Table 1 shows the relationship between heater voltage and heat flux. The two heater voltages used for the evaluation of the device were 70 and 85 volts. All

Table 1. Heater voltage and heating flux

Voltage	Flux, Cal/min
65	32,000
70	43,700
75	49,500
80	56,900
85	70,000
90	78,200

heaters were operated at the same voltage during the evaluation.

Another series of runs was made to determine the heat transfer coefficient for vapor condensing along the top of the tube. For these runs the tube was horizontal and the reboiler connected. The bottom of the tube was filled with acetone to a level of about one-eighth inch. This level was just below the level required for the siphon to measure the flow back to the reboiler. This layer of acetone was heated just to the boiling point so condensation of the vapor from the reboiler would not take place along this surface.

Cooling water was flowing in the channel along the top of the tube. The reboiler was turned on to a high enough heat flux so that vapor was flowing to

the condenser, thus assuring that condensation was taking place along the entire tube.

The amount of acetone flowing through the siphon from the tube to the reboiler was the amount of acetone condensed along the tube. An independent measure of the quantity was obtained from the energy balance of the cooling water. From this data a value for the overall heat transfer coefficient along the top of the tube was calculated to be 47.5 Btu/ft^2 ^{o}F hr. This value agrees well with published heat transfer coefficients (6).

During the operation of the column there was considerable heat loss by conduction from the heaters to the cooling water through the column walls. Therefore, the calculations of the heat input and heat output must be modified to take the conduction losses into account. Q_c will denote the actual heat gained by the cooling water due to condensation and Q_v will denote the actual heat supplied for vaporization by the heaters. These are total values and do not represent how these values vary with position along the tube.

The number of theoretical stages was calculated by the McCabe-Thiele (6) procedure. The assumption of equal molal overflow required by this procedure was not met in the horizontal tube, therefore the data must be interpreted to mean the equivalant number of McCabe-

Table 2. Results of runs at total reflux

Run No.	Tilt	Q_V, cal/min	Q_C, cal/min	Stages*
ALT	3°	27,300	13,900	1.8
AHT	3°	43,200	13,900	1.8
BLT	3°	27,300	17,200	2.3
BHT	3°	43,200	17,200	2.1
BLH	Horz	27,300	17,200	2.5
BHH	Horz	43,200	17,200	2.2
CLH	Horz	27,300	20,700	3.1
DHH	Horz	43,200	24,900	3.5

*The reboiler stage is not included.

Thiele stages to effect the separation observed.

Table 2 shows the results of the total reflux runs. Runs ALT, AHT, BLT, and BHT were duplicated two times. Run BLH was repeated three times. The average data are shown in Table 2. The reproducibility of the number of stages obtained was quite good, within approximately 0.1 stage.

Table 3 shows the results at partial reflux. The reflux at the condenser end was held nominally at 2.0, but this was difficult to measure as accurately as the L/V ratio at the reboiler end of the column. Therefore, the equivalant number of McCabe-Thiele stages was calculated

Table 3. Results of runs made at partial reflux

Run No.	L/V*	Tilt	Q_v, cal/min	Q_c, cal/min	Stages**
4	1.7	3°	27,300	14,700	2.3
7	1.7	3°	27,300	20,500	2.9
12	1.8	3°	43,200	15,300	1.0
15	1.6	3°	43,200	18,300	4.2
20	1.3	Horz	27,300	15,600	2.9
23	1.6	Horz	27,300	19,800	4.5
28	1.4	Horz	43,200	14,900	1.9
31	2.2	Horz	43,200	26,100	5.4

* L/V is calculated at the reboiler end of the tube.
**The reboiler stage is not included.

based on the L/V ratio at the reboiler end. None of these runs was duplicated.

A summary of the concentrations involved in the various runs is shown in Appendix.

Effect of Heat Input-Output Ratio

The effect of the heat input-heat output ratio along the tube is very important in the performance of the horizontal distillation device. R is defined as the ratio between Q_v and Q_c (i.e., $R = Q_v/Q_c$). The plots

Figure 3. The relationship between the number of stages and the heat input-output ratio, R, at total reflux

High Q_V = 43,200 cal/min

Low Q_V = 27,300 cal/min

The reboiler stage is not included.

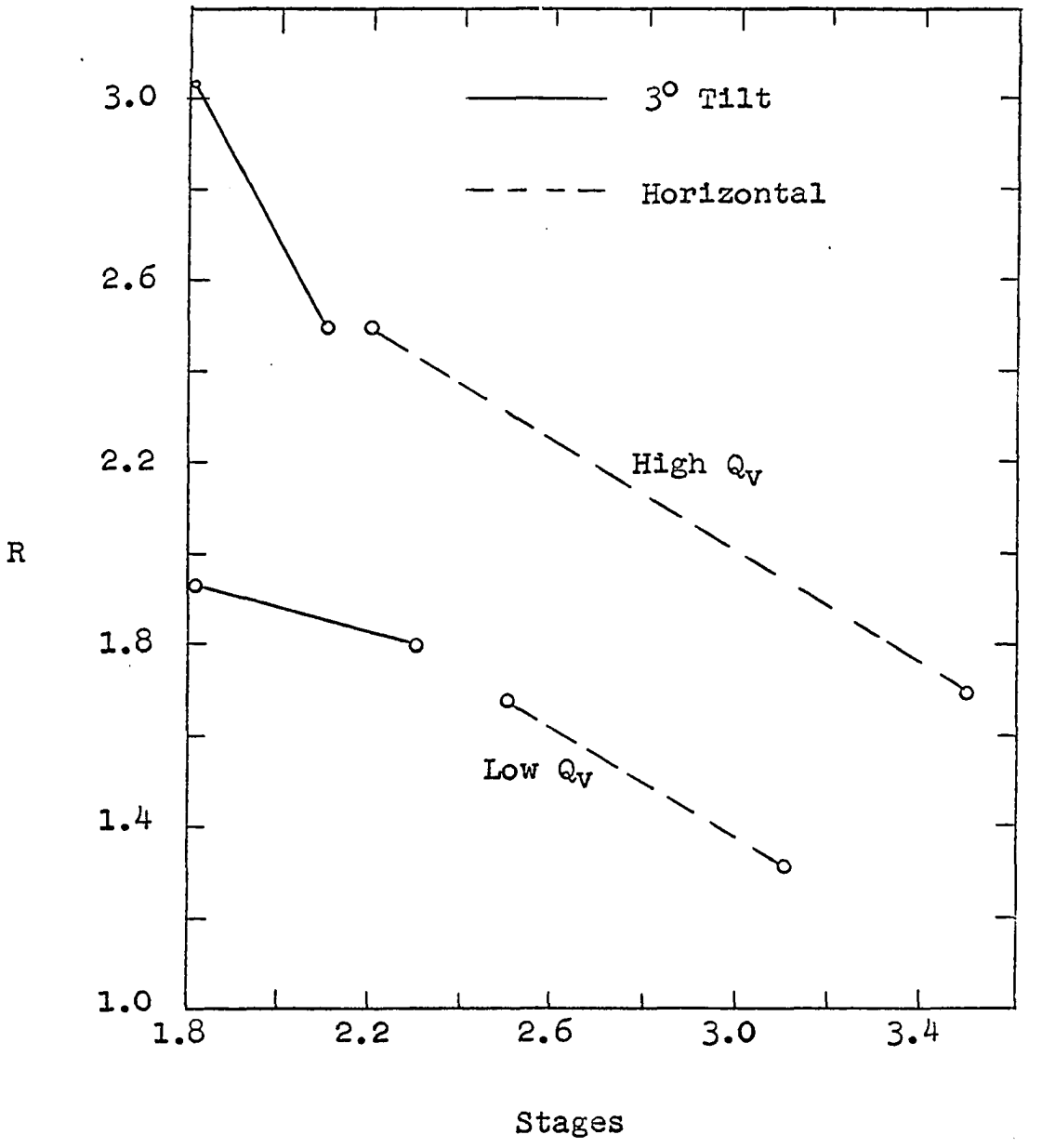
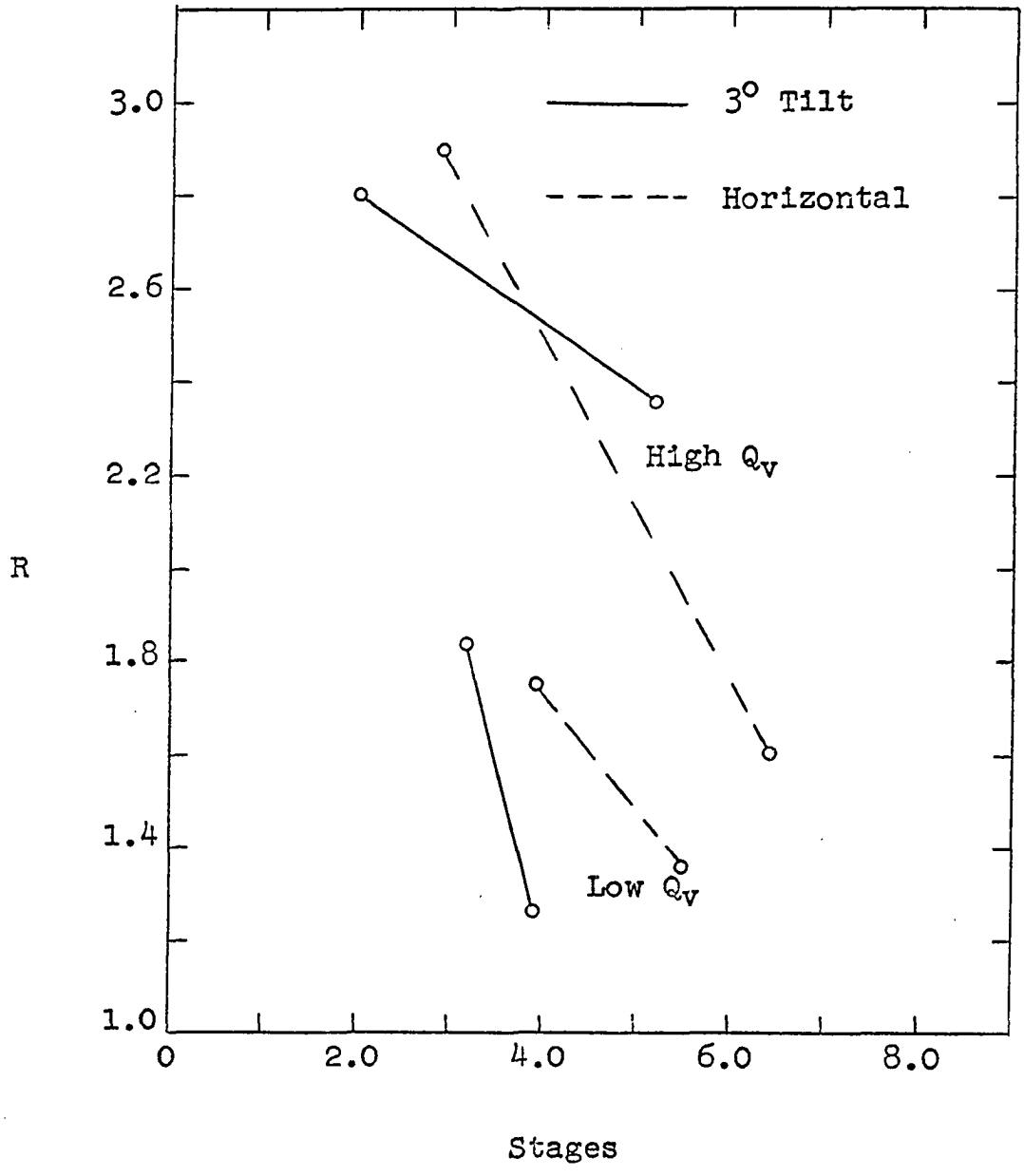

3° Tilt
Horizontal
High Q_V
Low Q_V
R
3.0
2.6
2.2
1.8
1.4
1.0
1.8
2.2
2.6
3.0
3.4
Stages

Figure 4. The relationship between the number of stages and the heat input-output ratio, R, at partial reflux

High Q_V = 43,200 cal/min

Low Q_V = 27,300 cal/min

The reboiler stage is not included.


3° Tilt
Horizontal
High Q_v
Low Q_v
R
3.0
2.6
2.2
1.8
1.4
1.0
0
2.0
4.0
6.0
8.0
Stages

of this ratio versus the equivalant McCabe-Thiele stages using heat input as a varying parameter are shown in Figures 3 and 4. Figure 3 shows the runs at total reflux and Figure 4 shows the runs at partial reflux. In each plot the stage for the reboiler has been subtracted from the total number of stages.

Examination of Figure 3 indicated that the lower R was, the better the separation. Figure 4 indicated much the same thing, but the data were not reproduced, therefore, the plot may not be as accurate as Figure 3. It should be pointed out that the separation was not only a function of R, but also of the value of Q_v. If the plots were extrapolated to an R value equal to one, the higher Q_v would give the greatest number of stages.

Figures 3 and 4 do not indicate the full importance of the heat input-output ratio. R was not a constant, but varied with position along the tube. The heat input was uniform along the entire column, but the heat output varied because the cooling water temperature rose as the water moved from the condenser end to the reboiler end. Thus, the heat input-output ratio was close to one at the condenser end and increased as the water traveled to the reboiler end of the tube. Therefore, Figures 3 and 4 indicate only average values

of the heat input-output ratio.

The point values of R will be defined as r.

$$r = \frac{q_v}{q_c} \qquad (1)$$

where

q_v = heat input, cal/min/unit length of tube

q_c = heat output, cal/min/unit length of tube.

Figures 5, 6 and 7 indicate the importance of this r ratio. In these figures the r ratio was plotted against the position along the tube as well as the plot of vapor and liquid acetone concentration against the position along the tube.

Figure 5 is a plot of Run AHT which had a separation equivalant to 1.8 McCabe-Thiele stages. The r ratio was uniform at a high value for about one-third the tube length and then dropped rapidly for the remaining tube length to a final value of about 1.5. When the r ratio was high it was observed that the vapor and liquid concentrations did not increase as the vapor traveled from the reboiler to the condenser. It was desirable to have these concentrations increase with position to get a high degree of separation. This increase in concentration was noted as the r ratio was lowered. This plot also indicates that the r

Figure 5. Plot of vapor and liquid concentration profile data and heat input-output ratio, r, profile data for Run AHT

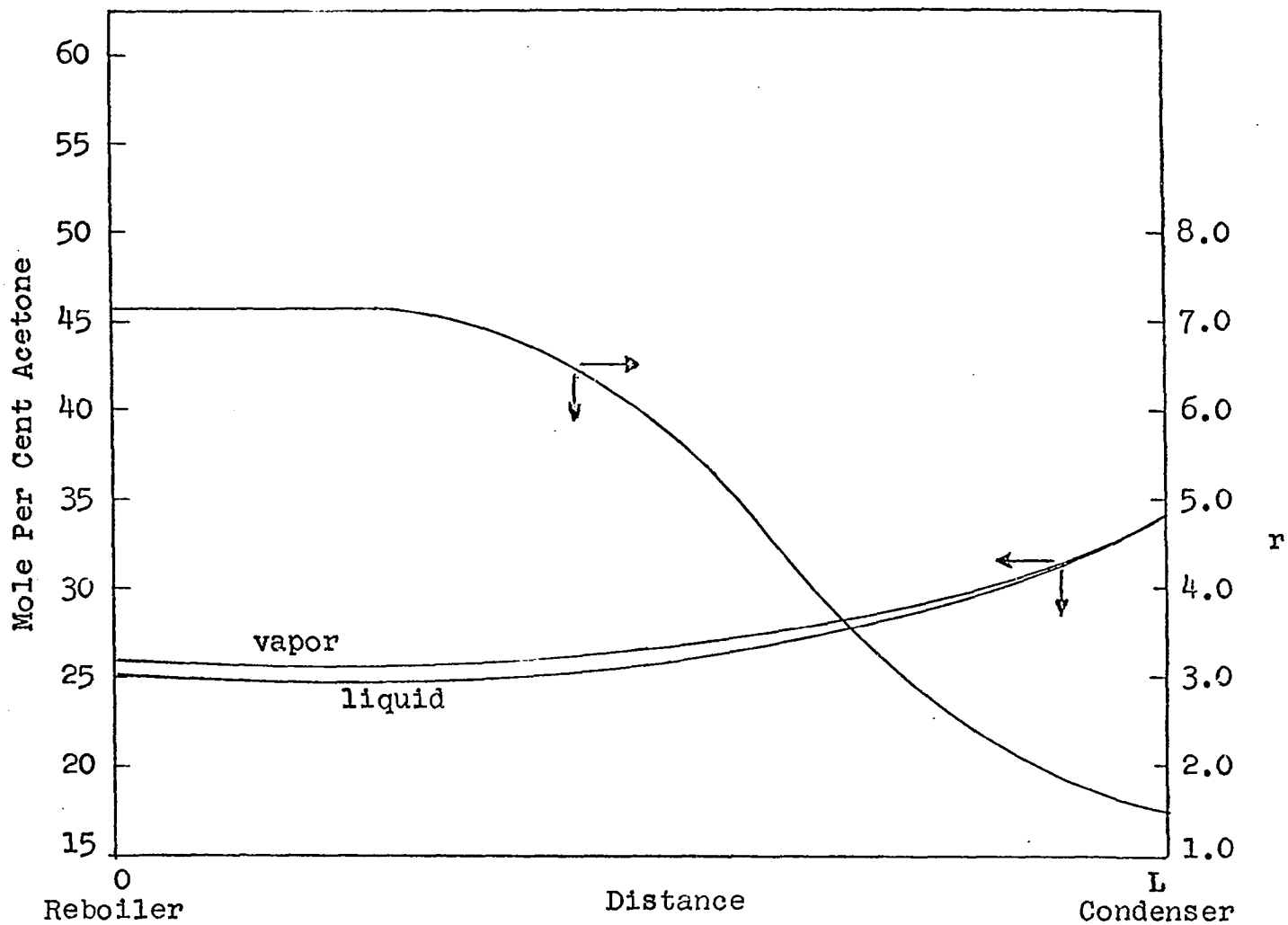

Mole Per Cent Acetone
60
55
50
45
40
35
30
25
20
15
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
r
vapor
liquid
0
Reboiler
Distance
L
Condenser

Figure 6. Plot of vapor and liquid concentration profile data and heat input-output ratio, r, profile data for Run BHH

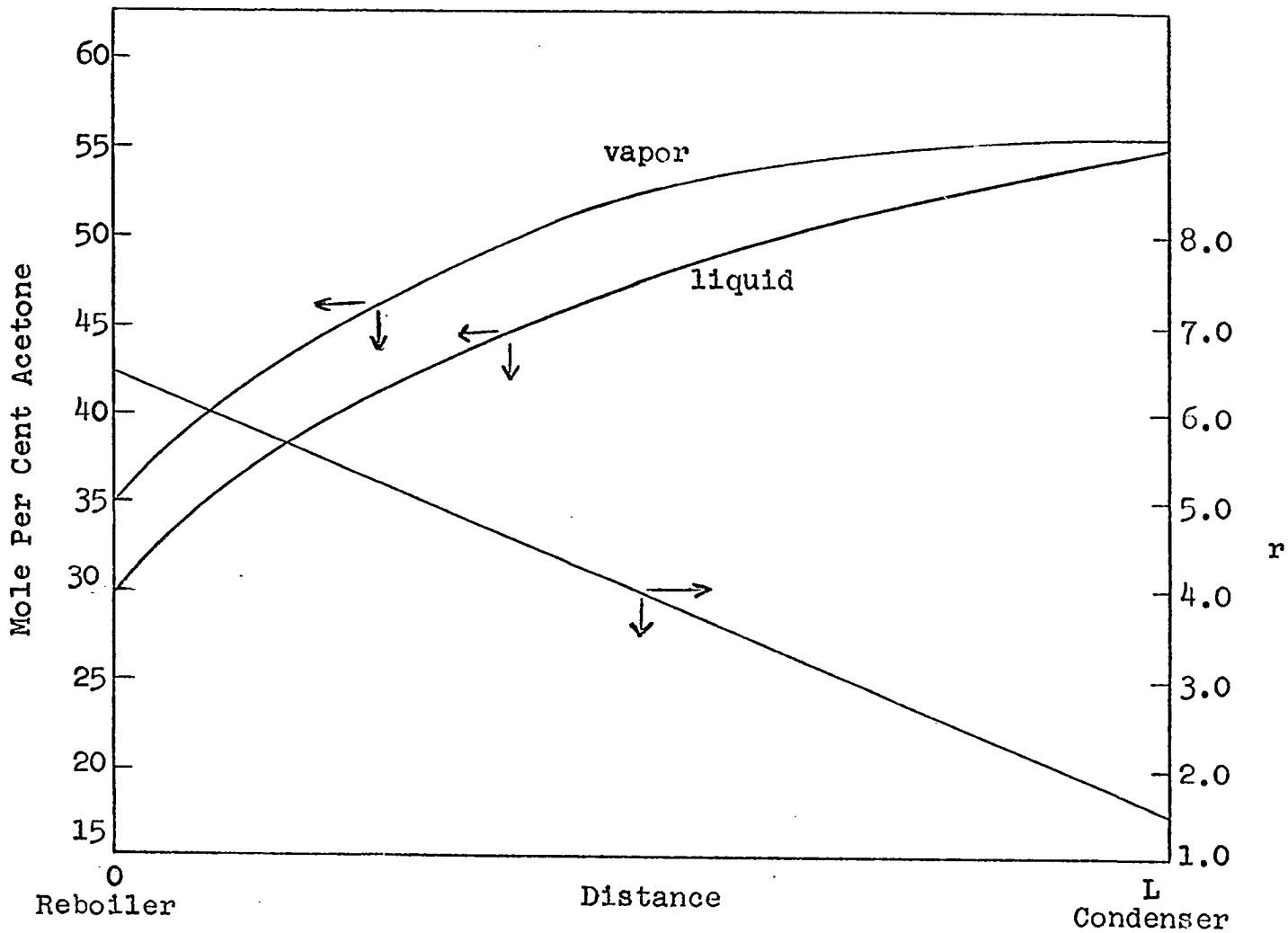
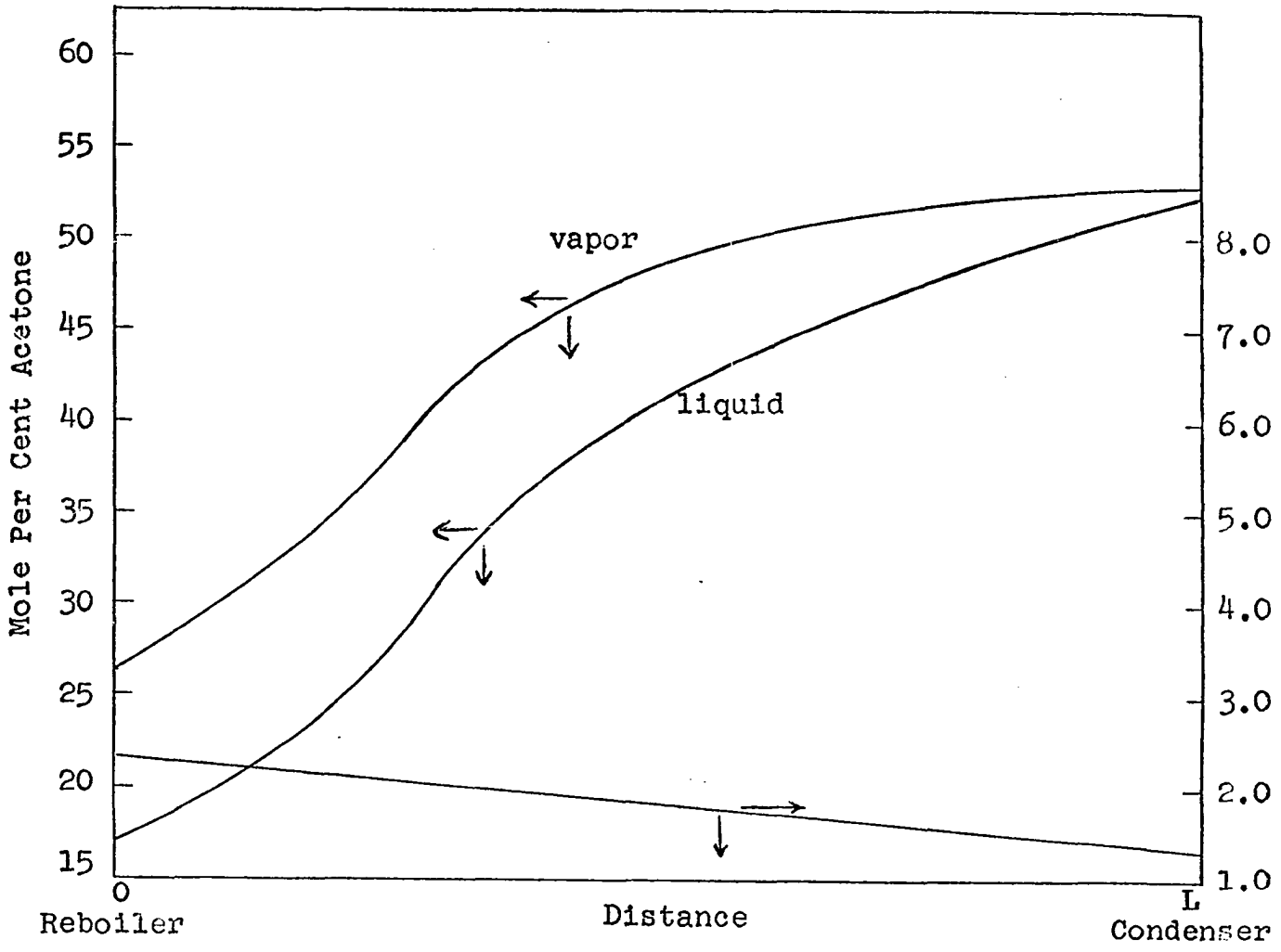

60
55
50
45
40
35
30
25
20
15
Mole Per Cent Acetone
vapor
liquid
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
r
0
Reboiler
Distance
L
Condenser

Figure 7. Plot of vapor and liquid concentration profile data and heat input-output ratio, r, profile data for Run DHH


60
55
50
45
40
35
30
25
20
15
Mole Per Cent Acetone
vapor
liquid
8.0
7.0
6.0
5.0
4.0
3.0
2.0
1.0
r
0
Reboiler
Distance
L
Condenser

ratio should be low to get a good separation.

Figure 6 is a plot of Run BHH which had a separation equivalant to 2.3 McCabe-Thiele stages. For this run the r ratio decreased linearly with position. Also the r value at the reboiler end of the tube was not as large as in Figure 5. The vapor and liquid concentrations increased with position, which was desired. It was important to note the separation between the vapor and liquid lines at any given distance. If the separation between liquid and vapor lines was large, a good separation was achieved in the distillation device.

Figure 7 is a plot of Run DHH which had a separation equivalant to 3.5 McCabe-Thiele stages. For this run the r ratio did not change very much with position and was at a low value. The vapor and liquid acetone concentration increased rapidly with position and the separation between the liquid and vapor lines was large. These observations indicate that such conditions are favorable for good separation between the low and high boiling compounds.

Examination of this profile data adds additional insight into the nature of the heat input-output ratio. R should be close to one as possible and should be constant for a good separation. If the R ratio is less than one a good separation will be achieved, but the

capacity of the distillation column will be reduced.

Effect of Tilt

The separation achieved in the horizontal distillation device depends upon the tilt of the tube as well as the heat input-output ratio.

A statistical comparison of Runs BLT, BHT, BLH and BHH indicated there was a significant improvement (at the 0.90 significance level) in the separation achieved by horizontal operation over that for a 3^{o} inclined position. For the case of partial reflux the data were much the same, but these runs were not duplicated, therefore, the data may not be as accurate as the total reflux runs. The separation was better in the horizontal position because the liquid holdup was larger in this position. The larger holdup resulted in an increase in the number of times the vapor from the reboiler condensed and reboiled during its journey through the tube.

One important aspect of the operation should not be overlooked, namely operation is easier in the horizontal position. In the horizontal position all liquid samples were easier to take because a pool of liquid exists at all points in the tube. When the tube was inclined an insufficient liquid pool was present to obtain good samples near the condenser end. Therefore,

the liquid profile data were more accurate when the device was operated in a horizontal position.

Pressure Drop Measurements

One of the most important advantages of a horizontal distillation column is the very low pressure drop of the device. The pressure drop was so low it was very difficult to get an accurate indication of it. Several different methods were used. The best was a simple manometer connected across the column.

Pressure drops of less than 5 mm water were observed for the device. There did not seem to be any significant difference in the pressure drop for horizontal or inclined operation, nor was any difference observed for the different heat input-heat output ratios.

Eberlin (2) reports pressure drops of about one mm water per theoretical stage for a small glass column. This seems to be in good agreement with the data observed in this experiment.

The pressure drops observed in this experiment were far less than would be expected in normal tray-type distillation towers or even as reported for other types of horizontal devices (5, 8). This small pressure drop was expected in a horizontal distillation device and its proof leads to visions of an important

use of such a fractionator, namely distillation under very high vacuum.

Comparison with Tray Type Columns

An advantage of the horizontal distillation column is the low pressure drop across the tube. As a result of the equipment design to obtain this low pressure drop, the heat input to the horizontal device was slightly larger than the heat input required to carry out the same separation in a conventional tray type vertical column.

A comparison of the horizontal distillation device and a conventional tray type column was made using data from the partial reflux Runs 12 and 28. The separation achieved in these Runs is shown in the Appendix. The comparison was made using the same energy input, production rate (distillate rate) and separation in the horizontal tube and the conventional column.

The production rate for Run 12 was 3.50 gram moles per minute. This production rate was obtained at a total heat input of 62,000 calories per minute including the reboiler and column heaters and an external reflux ratio of 0.75. To obtain the same production rate and separation in a conventional tray type distillation column at the same total heat input would require a reflux ratio of 1.36 in a one stage column.

For Run 28, the product on rate was 3.66 gram moles per minute, total heat input was 62,000 calories per minute and a reflux of 0.80. The required reflux ratio for a conventional 1.7 plate column was 1.20.

The results of the comparison indicate that the energy requirements are not greatly different for equivalent separations. The horizontal distillation apparatus requires a slightly lower reflux ratio than a conventional tray type column for a given set of conditions. Because the reflux ratios are very close for a given separation, the operating expenses on the basis of energy input should not be much different for a horizontal device or a conventional distillation tower.

MATHEMATICAL DEVELOPMENT

Derivation

A mathematical model was developed to express the liquid concentration profile as a function of the parameters of the system. This model is based on operation of the distillation device in a horizontal position.

The following assumptions were made:

1. Operation at steady state.
2. Thin liquid film along the tube.
3. No concentration gradient with respect to the width or height of liquid film.
4. No concentration profile in the vapor stream with respect to the height or width of the tube.
5. No mass transfer between the liquid drops falling from the cold surface and the passing vapor stream.
6. Vapor and liquid at equilibrum along the bottom of the tube.
7. Vapor condensation along the entire upper surface of the tube.
8. Uniform condensing rate with respect to tube length.

9. Uniform heat input-output ratio along the length of the tube.
10. Constant molal heat of vaporization.
11. Constant slope of the molal vapor-liquid equilibrum curve.

The differential element selected for the material and energy balances used to develop the mathematical model for concentration profiles is shown in Figure 8. The vapor stream, v, leaving the element was defined as follows:

$$v = \frac{q_v}{\lambda} \tag{2}$$

where

q_v = heat supplied to the bottom of the tube, cal/min/in

λ = heat of vaporization, cal/gram mole

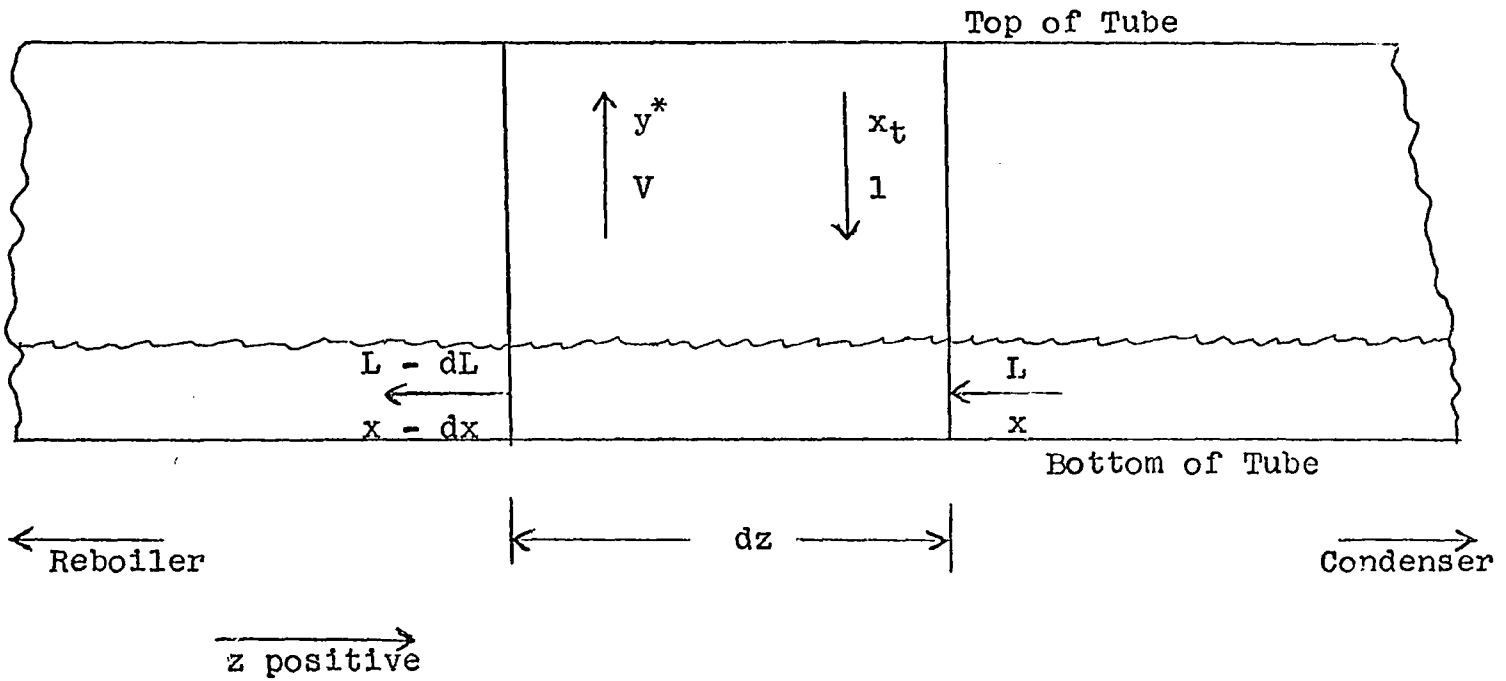
and the liquid, l, entering the element from condensation was

$$l = \frac{q_c}{\lambda} \tag{3}$$

where

q_c = heat removed by condensation, cal/min/in.

Figure 8. Identification of the element selected for use in mathematical development


Top of Tube
y*
V
x_t
l
L - dL
x - dx
L
x
Bottom of Tube
dz
Reboiler
Condenser
z positive

The overall material and energy balance became

$$\frac{dL}{dz} = \frac{q_v}{\lambda} - \frac{q_c}{\lambda} \qquad (4)$$

where

L = flow of liquid along the bottom of the tube, gram moles/min

z = unit length of the tube, inches.

The component material and energy balances were combined

$$\lambda L\frac{dx}{dz} + \lambda x\frac{dL}{dz} = q_v y^* - q_c x_t \qquad (5)$$

where

x = mole fraction of the most volatile component in the liquid

y^* = mole fraction of the most volatile component in the vapor which is in equilibrum with the liquid

x_t = mole fraction of the most volatile component in the liquid condensed along the top of the tube.

Let

$$r = \frac{q_v}{q_c} \tag{6}$$

and

$$y^* = mx \tag{7}$$

where

m = slope of the vapor-liquid equilibrum curve.

The concentration of the liquid drop, x_t, was actually the vapor concentration from some point upstream from the chosen differential element. This distance was d inches. Thus

$$x_t = mx_d \tag{8}$$

where

x_d = concentration of the liquid d inches upstream from the location of x_t.

Combining the above three equations with the component material and energy balance, equation 5 and substution for dL/dz from equation 4 yielded

$$\frac{\lambda L}{mq_c}\frac{dx}{dz} - \left[\frac{1 - r + mr}{m}\right] x = -x_d \tag{9}$$

The right hand side of the above equation, x_d, was a function of z and distance, d, upstream where the vapor condensed in the element was vaporized from the liquid stream. Thus

$$x_d = f(z - d). \tag{10}$$

The above equation was expanded in a Taylor's series using only the first three terms of the expression,

$$x_d = f(z) - f'(z)d + f''(z)\frac{d^2}{2} \tag{11}$$

where

$$f(z) = z.$$

Substution of equation 11 in equation 9 and rearranging,

$$\frac{d^2}{2}\ \frac{d^2x}{dz^2} + \left[\frac{\lambda L}{mq_c} - d\right]\frac{dx}{dz} + \left[1 - \left(\frac{1 - r + mr}{m}\right)\right] = 0 \tag{12}$$

The above equation was put in dimensionless form by letting

$$s = \frac{z}{D} \tag{13}$$

where

D = total length of the tube, inches.

Equation 12 became

$$\frac{d^2}{2D^2}\frac{d^2x}{ds^2} + \left[\frac{\lambda L}{Dmq_c} - \frac{d}{D}\right]\frac{dx}{ds} + \left[1 - \left(\frac{1 - r + mr}{m}\right)\right]x = 0 \tag{14}$$

Let

$$\alpha = \frac{d}{D} \tag{15}$$

$$\beta = \frac{\lambda L}{Dmq_c} \tag{16}$$

$$\gamma = \frac{1 - r + mr}{m} \tag{17}$$

Simplified, Equation 14 was then,

$$\frac{\alpha^2}{2}\frac{d^2x}{ds^2} + \left[\beta - \alpha\right]\frac{dx}{ds} + \left[1 - \gamma\right]x = 0 \tag{18}$$

The solution of the above equation required two boundry conditions. The boundry conditions chosen were,

$$\begin{aligned} &\text{at} \quad s = 0, \quad x = x_1 \\ &\qquad s = 1, \quad x = x_2 \end{aligned} \tag{19}$$

At the condenser end of the tube s = 1, therefore x_2 was the distillate composition. At the reboiler end of the tube s = 0, thus x_1 was the composition of the liquid discharged to the reboiler.

Integration of equation 18 and application of the boundry conditions resulted in the desired equation relating the liquid composition to location along the tube,

$$x = \left[\frac{x_2 - x_1 e^{(A - B)}}{e^{(A + B)} - e^{(A - B)}}\right] e^{(A + B)s} - \left[\frac{x_2 - x_1 e^{(A + B)}}{e^{(A + B)} - e^{(A - B)}}\right] e^{(A - B)s} \tag{20}$$

where

$$A = \frac{1}{\alpha^2}\left[\alpha - \beta\right]$$

$$B = \frac{1}{\alpha^2}\left[\left[\alpha - \beta\right]^2 + 2\alpha^2 (\gamma - 1)\right]^{\frac{1}{2}}.$$

It was felt that desired operation of the distillation device should be when the value of r is equal to one. For this case γ is equal to one and equation 20 reduces to,

$$x = \left[\frac{x_2 - x_1}{e^{2A} - 1}\right] e^{2As} - \left[\frac{x_2 - x_1 e^{2A}}{e^{2A} - 1}\right] \tag{21}$$

where

$$A = \frac{1}{\alpha^2}\left[\alpha - \beta\right].$$

Discussion

The model was tested by comparing the predicted concentration profile obtained from the model with the observed profile in the tube. The restrictions on the model that q_c and r are independent of position were difficult to achieve in the experimental device. This was because the cooling medium along the top of the tube could not be kept at a uniform temperature. However, Run DHH did approximate the assumptions used and its concentration profile was compared with the mathematical model.

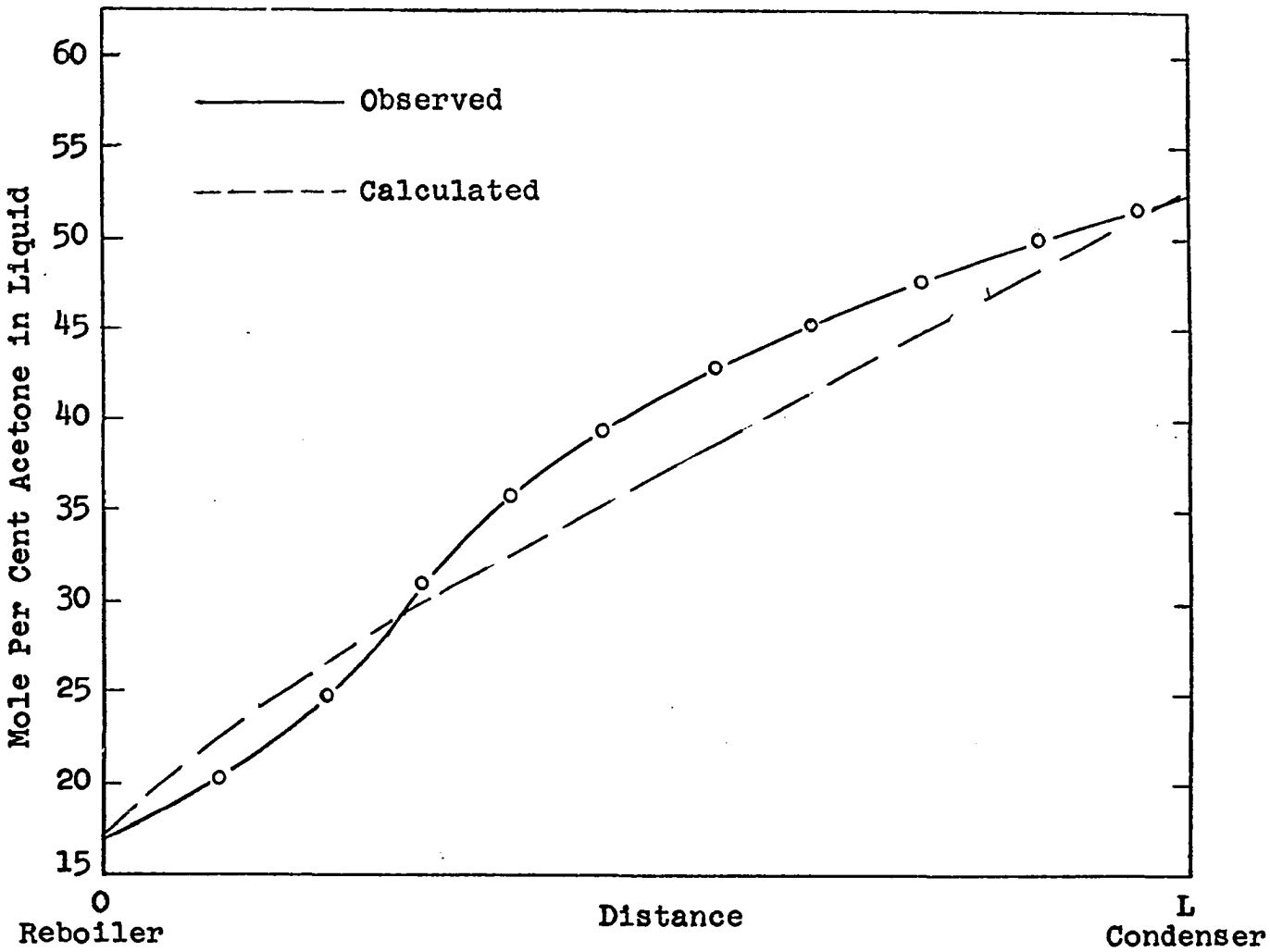
A trial and error technique was used to find α. The value of β was calculated to be 0.45 and γ was 1.16. Figure 9 shows the results of this trial and error fit.

Figure 9. Comparison of the concentration profile for Run DHH and the profile predicted from the model

$\alpha = 0.3$

$\beta = 0.45$

$\gamma = 1.16$


Observed
Calculated
Mole Per Cent Acetone in Liquid
60
55
50
45
40
35
30
25
20
15
0
Reboiler
Distance
L
Condenser

The best fit was obtained when α was 0.3. The fit was not exact because the assumptions were not all met in the experiment. At locations where s is less than 0.3 the fit was not expected to be good because in this region the condensing liquid was vaporized in the reboiler instead of the tube as assumed in the model.

A sensistivity analysis was made of the model using the data from Run DHH. From this analysis it was determined that if the value of β was changed plus or minus ten per cent, the profile was not much different than the one plotted in Figure 9. The β factor contains the slope of the equilibrum line, the molal heat of vaporization, and the amount externally refluxed as well as the heat removal rate. If the value of γ was changed plus or minus ten per cent the profile changed appreciably. The γ factor contains the slope of the equilibrum curve and the heat input-output ratio r.

The results of the sensistivity analysis indicate that the separation achieved is dependent more on the r ratio than on the heat removal rate or the amount externally refluxed.

Two general observations about the model can be made. First, the separation depends to a considerable extent on the value of r as well as on q_c. Second,

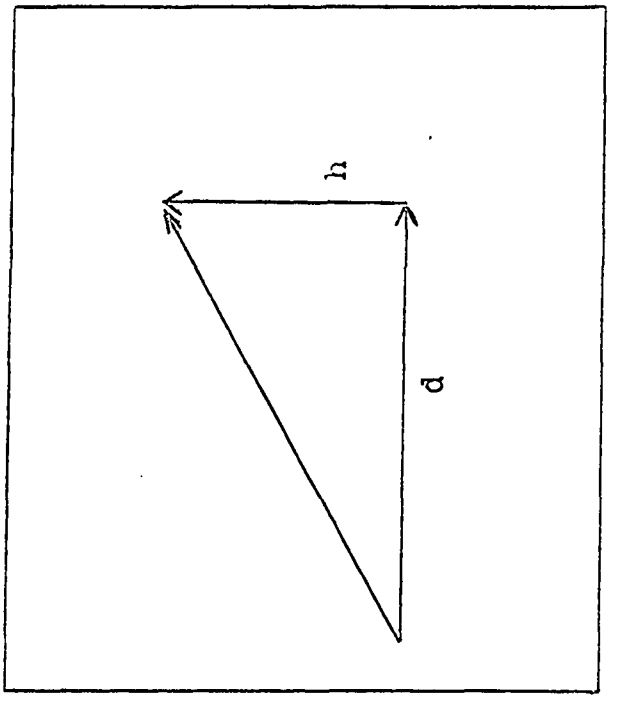
the closer r is the unity the better the separation. Both of these observations were consistent with the observed experimental data as shown in Figures 3 and 4.

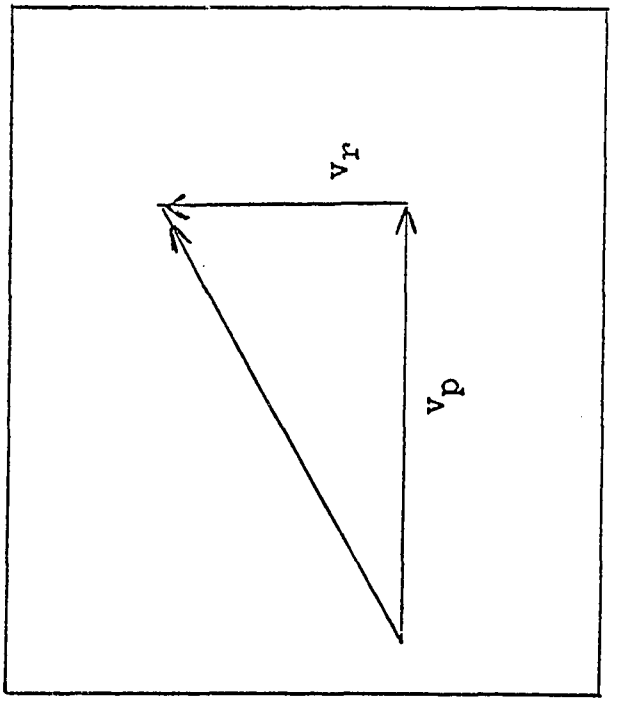
Alpha was defined as d/D, where D is the total length of the tube. The linear distance d is that horizontal distance which the vapor travels after it has left the liquid on the bottom of the tube before it condenses. This distance is related to the horizontal velocity due to the vapors entering from the reboiler and the vertical velocity of the vapors leaving the liquid pool along the tube. Both of these velocities depend on the geometry of the tube as well as the boiling rates in the reboiler and the tube.

If it is assumed that these velocities are in layer plug flow it is possible to calculate the value of d. The two velocities are shown in vector form in Figure 10. The hypotenuse describes the path velocity of the vapor as it boils off the bottom of the tube. The hypotenuse of the triangle shown in Figure 11 represents the distance the vapor travels and relates the distance to the tube height. The triangles shown in Figures 10 and 11 are similar, therefore the following relationship is obtained:

Figure 10. Velocity vectors

Figure 11. Distance triangle


h
d


v_r
v_p

$$d = \frac{v_p}{v_r} h \qquad (22)$$

where

v_p = vapor velocity from the reboiler

v_r = vapor velocity due to heat input along the tube

h = height of the tube.

For Run DHH, $v_p = 8.3$ ft/sec, $v_r = 0.56$ ft/sec and $h = 3$ inches. Thus the calculated value of d was 44 inches or $\alpha = 0.46$. The value of alpha calculated by fitting the model to the observed concentration profile for Run DHH was 0.3. The agreement was not good because the velocity of the vapor from the reboiler was in turbulent flow, thus the assumption of layer plug flow was not met.

If a "stage" is defined as the number of times the liquid is evaporated and condensed during its journey through the tube, then $1/\alpha$ is the number of stages in the tube. For Run DHH the number of stages, according to the above criteria, using the alpha obtained from the profile model was 3.33. This compares very well with the 3.5 equivalent number of McCabe-Thiele stages calculated.

The mathematical model derived for liquid concentration profiles in the tube is not limited to total reflux runs. The only requirement for partial reflux operation is that the flow rate to the condenser end of the tube be known. If the equation is to be used directly for partial reflux operation then the distillation column must be fed at the reboiler. If the column is center fed, the equation must be applied to both the stripping and rectification part of the column and the solutions to the equations matched at the feed point.

ACCOMPLISHMENTS AND CONCLUSIONS

1. A small scale horizontal distillation device was constructed and operated satisfactorily using the acetone-methanol system.

2. At total reflux the maximum separation achieved was equivalant to 3.5 McCabe-Thiele stages.

3. The pressure drop across the horizontal device was less than 5 mm water at all operating conditions tested.

4. A better separation was achieved in a horizontal position rather than tilted 3° to the reboiler end.

5. The separation obtained was primarily dependent on both the heat input-output ratio and the absolute value of the heat input. The best separation will probably be when the heat input-output ratio is approximately one.

6. A mathematical model for the liquid concentration profile along the tube was developed and successfully tested.

7. At total reflux the number of stages calculated from the model was in agreement with the number of stages calculated by the McCabe-Thiele method.

8. The vapor velocity from the reboiler combined with the column geometry has an important bearing on the degree of separation obtained.

RECOMMENDATIONS

1. The horizontal distillation device should be further tested on other systems.

2. The cooling system for the upper surface of the tube should be redesigned to allow the heat input-heat output ratio to be independent of the position along the tube.

3. A series of experiments should be made to determine how the vapor velocity from the reboiler affects the separation.

4. A number of different geometry columns should be tested to determine the effect of column geometry on separation.

5. The thickness of the liquid film along the tube should be investigated to determine its importance.

6. A simpler model should be developed. This would allow for a simpler analysis of the data.

7. The model should be tested using partial reflux conditions.

8. A better sampling method should be developed to determine the vapor concentration profile in the tube.

NOMENCLATURE

A	constant in profile equation, dimensionless
B	constant in profile equation, dimensionless
D	length of tube, inches
d	length of stage, inches
h	height of tube, inches
L	liquid fed to tube, moles/min
l	liquid condensed along the top of the tube, moles/min
m	slope of equilibrum line
Q_c	total heat removed by condensation along the top of the tube, cal/min
q_c	heat removed by condensation along the top of the tube, cal/min/in
Q_v	total heat supplied to the bottom of the tube, cal/min
q_v	heat supplied to the tube, cal/min/in
R	ratio of total heat input to heat output, dimensionless
r	ratio of the point value of the heat input to heat output, dimensionless
s	dimensionless distance along the tube
v_p	velocity of the vapors from the reboiler, ft/sec
v_r	velocity of the vapors from the bottom of the tube, ft/sec
v	vapor leaving the bottom of the tube, moles/min
V	vapor rate, moles/min
x	mole fraction acetone in liquid, dimensionless
x_t	mole fraction acetone in drops condensed, dimensionless

x_d mole fraction acetone in liquid d inches upstream from the point of interest, dimensionless

y^* mole fraction acetone in equilibrum with a liquid concentration, dimensionless

z length, inches

α dimensionless constant in profile equation

β dimensionless constant in profile equation

γ dimensionless constant in profile equation

λ heat of vaporization, cal/gram mole

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Special acknowledgement goes also to my wife, Pat. Her patience during the many hours away from home is more than I am sure I could have endured.

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APPENDIX

Table A-1 shows the tabulation of the experimental data for determination of the equivalant McCabe-Thiele stages.

Table A-1. Experimental data

Run No.	L/V*	x_D	x_P	x_F	Stages
ALT	1	0.47	0.12	-	3.0
	1	0.44	0.14	-	2.5
AHT	1	0.38	0.07	-	3.0
	1	0.34	0.06	-	2.6
BLT	1	0.44	0.08	-	3.3
	1	0.54	0.17	-	3.3
BHT	1	0.41	0.09	-	3.0
	1	0.36	0.04	-	3.2
BLH	1	0.58	0.21	-	3.4
	1	0.55	0.14	-	3.6
	1	0.63	0.28	-	3.5
BHH	1	0.56	0.20	-	3.2
CLH	1	0.63	0.20	-	4.1
DHH	1	0.63	0.16	-	4.5
4	1.7	0.49	0.14	0.18	3.3
7	1.7	0.62	0.26	0.31	3.9
12	1.8	0.48	0.26	0.43	2.0
15	1.6	0.57	0.27	0.43	5.2
20	1.3	0.62	0.26	0.42	3.9
23	1.6	0.67	0.33	0.42	5.5
28	1.4	0.52	0.26	0.42	2.9
31	2.2	0.64	0.33	0.42	6.4

*Based on the lower portion of the column, see text.


65
468